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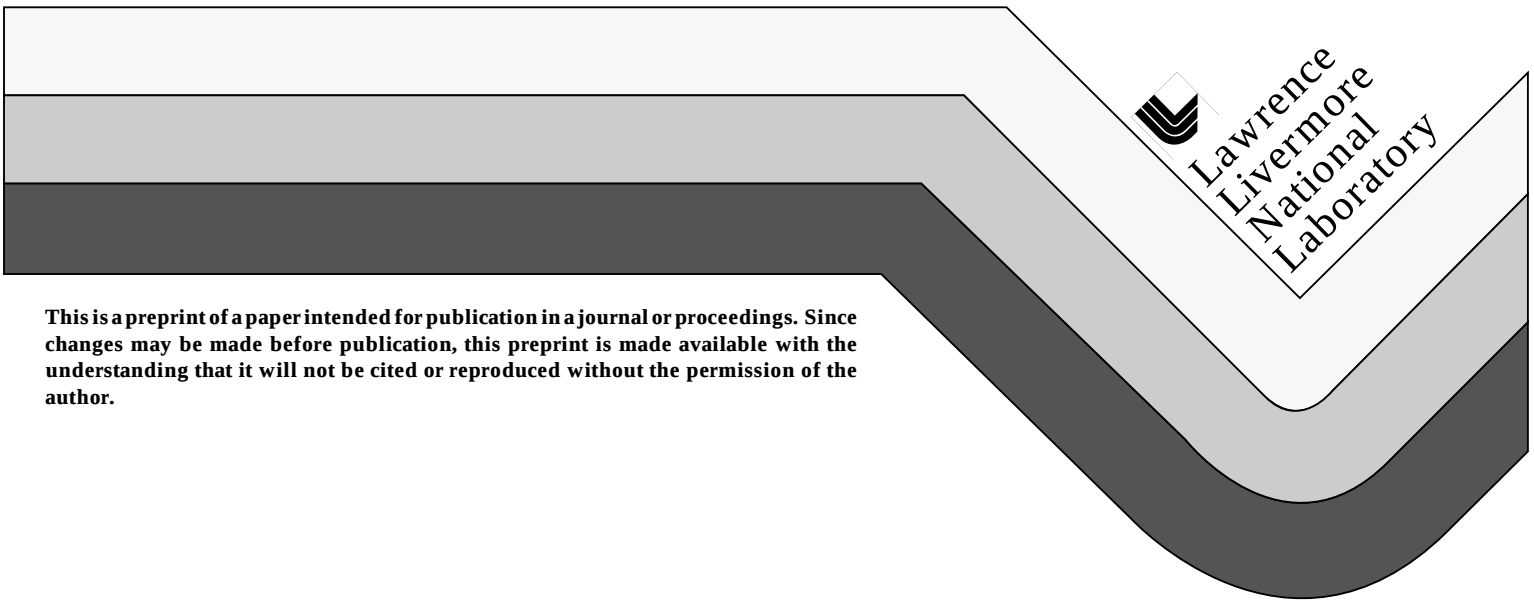

NO_X REDUCTION BY COMPACT ELECTRON BEAM PROCESSING

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Among the new methods being investigated for the post-combustion removal of nitrogen oxides (NO_x) are based on non-thermal plasmas. These plasmas can be produced by electrical discharge methods or electron beam irradiation. The application of electron beam irradiation for NO_x removal in power plant flue gases has been investigated since the early 1970's in both laboratory- and pilot-scale experiments. Electrical discharge methods are relatively new entrants in the field of flue gas cleanup. Pulsed corona and dielectric-barrier discharge techniques are two of the more commonly used electrical discharge methods for producing non-thermal plasmas at atmospheric pressure.

There are basically two types of reactions responsible for the depletion of NO by non-thermal plasmas: oxidation and reduction (see Figure 1).

In power plant flue gas treatment applications, the purpose of the plasma is to oxidize NO. The plasma produces OH radicals that play the major role in the simultaneous oxidation of NO and SO_2 to their respective acids. The presence of SO_2 lowers the power requirement of the oxidation process by recycling the OH radicals (see Figure 2). The presence of O radicals provide additional oxidation of NO to NO_2; the latter is then further oxidized by OH radicals to nitric acid. The desired products, in the form of ammonium salts, are then obtained by mixing ammonia with the formed acids. Some form of scrubbing is required to collect the final products.

Our experiments show that the presence of hydrocarbons promotes only the oxidation of NO to NO_2, but not the reduction of NO to N_2 and O_2. The chain-oxidation process in the presence of hydrocarbons is shown in Figure 3. Thus the use of hydrocarbon additives may be advisable only for lowering the power consumption of the plasma process in stationary combustion sources for which scrubbing of byproducts is acceptable.

For mobile engine applications, it is very important to make a distinction between NO re-

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Oxidation:

$$e + O_2 \rightarrow e + O(^3P) + O(^3P)$$

$$e + O_2 \rightarrow e + O(^3P) + O(^1D)$$

$$O(^3P) + NO + M \rightarrow NO_2 + M$$

$$O(^1D) + H_2O \rightarrow OH + OH$$

$$OH + NO_2 \rightarrow HNO_3$$

Reduction:

$$e + N_2 \rightarrow e + N(^4S) + N(^4S)$$

$$e + N_2 \rightarrow e + N(^4S) + N(^2D)$$

$$N(^4S) + NO \rightarrow N_2 + O(^3P)$$

$$N(^2D) + NO \rightarrow N_2 + O(^3P)$$

Figure 1. There are basically two types of reactions responsible for the depletion of NO by non-thermal plasmas: oxidation and reduction. Because of the relatively low concentration of NO, the plasma processing always starts with electron-impact reactions with the background gas molecules. The relative amount of oxidized products (NO_2 and HNO_3) to reduced products (N_2) is determined largely by the average kinetic energy of the electrons and the composition of the background gas. In diesel engine exhaust gases, the presence of large amounts of O_2 (around 10%) and H_2O (around 5%) make it difficult to avoid the oxidation process. The excited nitrogen atoms, $N(^2D)$, can also react with O_2 to produce NO.

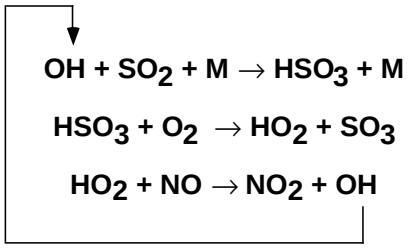
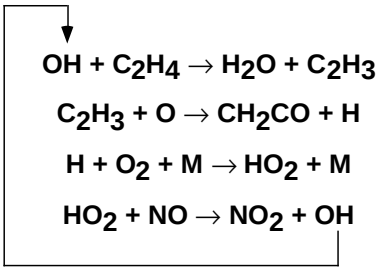


Figure 2. In flue gas treatment by non-thermal plasmas, the OH radical plays a key role in the simultaneous oxidation of NO and SO_2. The presence of SO_2 serves to lower the energy cost for oxidation of NO by converting OH to HO_2; the OH radical is then reproduced when NO is oxidized by HO_2.



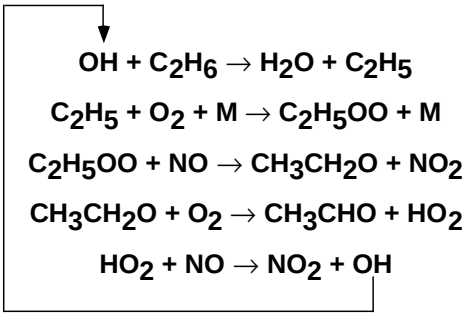


Figure 3. In non-thermal plasma processing at ambient temperature conditions, the presence of hydrocarbons enhances the oxidation of NO to NO_2, but not the reduction of NO to N_2 and O_2.

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moval by chemical oxidation and NO removal by chemical reduction. To avoid the need for scrubbing of process products, the desired method of NO removal is by chemical reduction; i.e. the conversion of NO to the benign products N_2 and O_2. For typical exhaust gases without additives, the only species that the plasma can produce to implement NO reduction is the N atom. From here on, the term “NO reduction” refers strictly to the reaction

$$N + NO \rightarrow N_2 + O.$$

The plasma produces N atoms through electron-impact dissociation of N_2 in the exhaust gas:

$$e + N_2 \rightarrow e + N + N.$$

The electron energy distribution in a plasma reactor is important because it determines the types of radicals produced in the plasma and the input electrical energy required to produce those radicals. Figures 4 and 5 show the cross sections for electron-impact reactions with N_2 and O_2. Note that large electron kinetic energies are required to optimize the dissociation of N_2 and the subsequent reduction of NO. Even with only 10% O_2 in the exhaust gas, the probability for dissociating O_2 is large compared to the probability for dissociating N_2 when the average kinetic energy of the electrons is small (less than 10 eV).

In discharge processing, the rate coefficients for electron-impact dissociation reactions strongly depend on the electron mean energy in the discharge plasma. In pulsed corona and dielectric-barrier discharge reactors, the non-thermal plasma is produced through the formation of statistically distributed microdischarges known as streamers. The electrons dissociate and ionize the background gas molecules within nanoseconds in the narrow channel formed by each microdischarge. The electron energy distribution in the plasma is complicated because the electric field is strongly non-uniform (e.g. because of strong space-charge field effects) and time dependent. During the microdischarge formation phase, the electron number rises drastically. Due to field strength enhancement in the ionization wave, the highest electron energies occur during this phase. The mean electron energy reaches values of more than 10 eV - suitable for large dissociation and ionization of the gas. However, since this is a highly transient phase, and since the ionization wave covers only small parts of the gap at the same time, this phase seems to be less important in producing most of the active radicals. Most of the species responsible for the

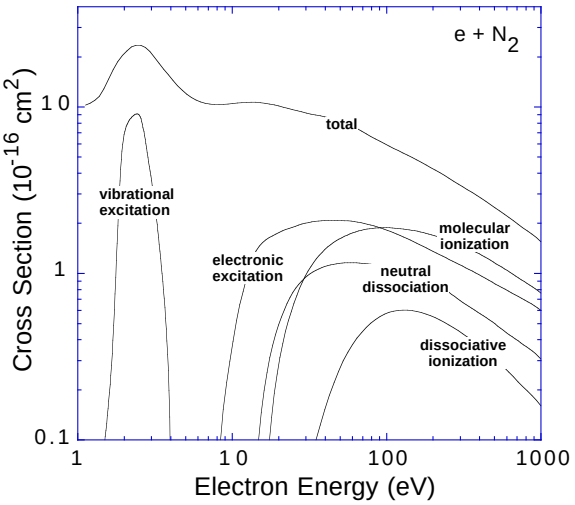

Figure 4. Cross sections for electron-impact reactions with N_2, shown as functions of the kinetic energy of an electron. The total collision cross section also includes contributions from processes not shown in the figure. The probability for a particular process is approximately equal to the ratio of the cross section for that process to the total collision cross section. When the average kinetic energy of the electrons in the plasma is small (less than 10 eV), a large fraction of the input electrical power is wasted in the vibrational excitation of N_2. Large electron kinetic energies are required to optimize the dissociation of N_2 and the subsequent reduction of NO.

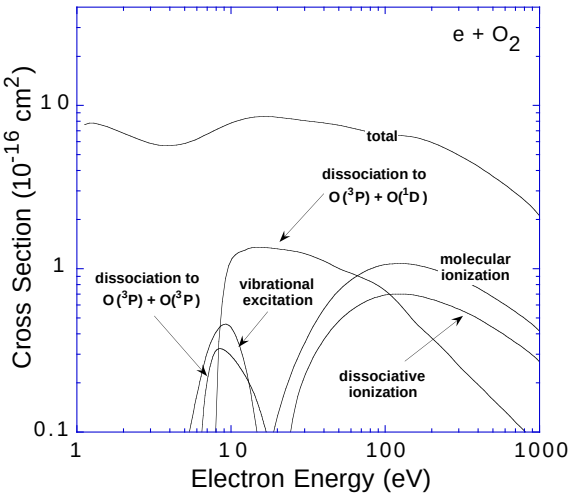

Figure 5. Cross sections for electron-impact reactions with O_2, shown as functions of the kinetic energy of an electron. The total collision cross section also includes contributions from processes not shown in the figure. Even with only 10% O_2 in the exhaust gas, the probability for dissociating O_2 is large compared to the probability for dissociating N_2 when the average kinetic energy of the electrons is small (less than 10 eV).

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chemical processing are generated in the microdischarge channels already established during the main current flow. In each microdischarge column, the electrons acquire a drift velocity, v_d, and an average energy corresponding to an effective E/n, i.e., the value of the electric field E divided by the total gas density n. The efficiency for a particular electron-impact process can be expressed in terms of the G-value (number of dissociation reactions per 100 eV of input energy) defined as

$$G\text{-value} = 100\ k / (v_d\ E/n)$$

where k is the rate coefficient (cm^3/molec-s). The rate coefficient k represents the number of reactions in a unit volume per unit time. The quantity v_d E/n represents the amount of energy expended by the electrons in a unit volume per unit time.

Under most conditions encountered in pulsed corona or dielectric-barrier discharge processing, the effective E/n is close to the value for breakdown (Paschen field) [1,2]. Figure 6 shows the breakdown values for the reduced field strength, E/n, as a function of the electrode gap spacing for an atmospheric pressure discharge. Note that for typical electrode gap spacings used in the implementation of pulsed corona or dielectric-barrier discharge reactors, the breakdown E/n is limited to values between 100 and 300 x 10^{-17} V-cm^2. For air-like mixtures, the effective E/n is around 150 x 10^{-17} V-cm^2, which corresponds to an electron mean energy of about 4 eV. The corresponding average electron energies are shown in Figure 7. There are two ways of increasing the electron mean energy: (1) use very narrow gap spacings (100 microns or less) to increase the breakdown E/

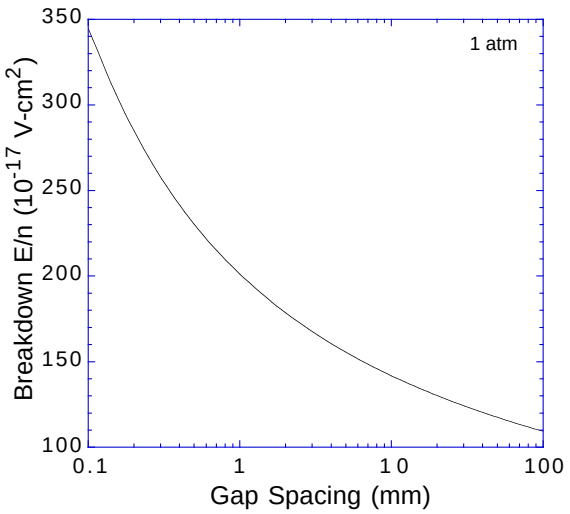

Figure 6. Breakdown values for the reduced field strength, E/n, as a function of the electrode gap spacing for an atmospheric pressure discharge. E is the electric field and n is the total gas density.

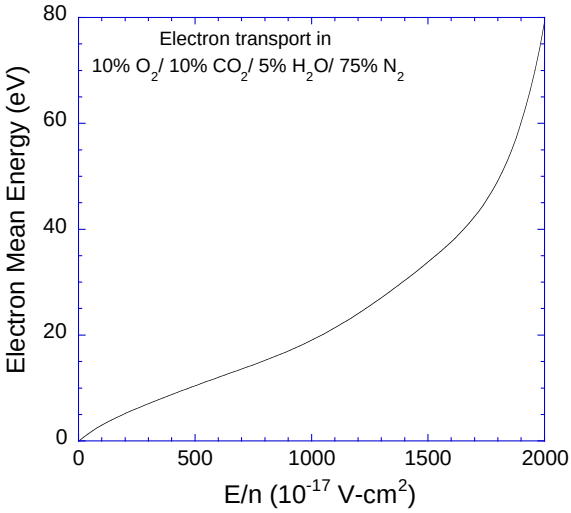

Figure 7. Average kinetic energy of the electrons as a function of the reduced field strength, E/n, in a gas mixture simulating a diesel engine exhaust. The E/n experienced by the plasma in electrical discharge reactors is typically less than 300 x 10^{-17} V-cm^2. The average electron kinetic energy is thus limited to values less than 10 eV.

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n for the same applied voltage, or (2) use very fast rising voltage pulses (10 nanoseconds or less risetime) to increase the breakdown E/n for typical gap spacings. We have investigated

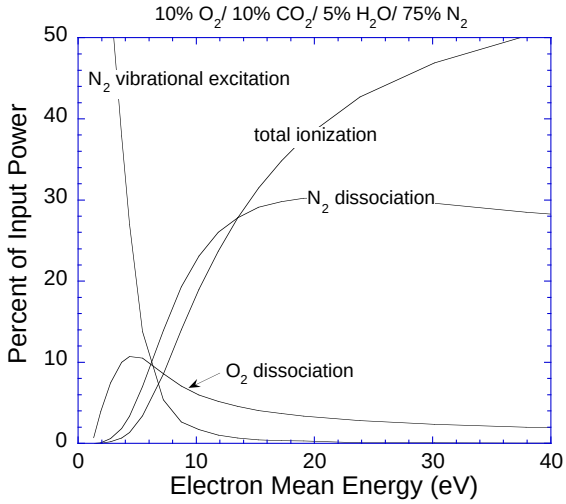

Figure 8. Dissipation of input electrical power as a function of the average kinetic energy of the electrons in a gas mixture simulating a diesel engine exhaust. There are other processes that dissipate the input power that are not shown in the figure. The electron mean energy in most electrical discharge reactors is typically between 3 to 6 eV. In this range, a large fraction of the input power is wasted in vibrational excitation of N_2 and a significant fraction goes into dissociation of O_2. The latter leads to significant oxidation of NO to NO_2 and subsequently nitric acid. High electron mean energies are required to optimize the dissociation of N_2, which leads to the chemical reduction of NO.

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both methods. In the first case we used a parallel-plate dielectric-barrier discharge reactor with a gap spacing of 50 microns. With such a narrow gap we have been able to increase the electron mean energy to around 10 eV, as verified by experiments using simple mixtures of NO or NO_2 in N_2. In the case of very fast rising voltage pulses, the breakdown E/n could increase to about twice the normal breakdown E/n, i.e. around 300 x 10^{-17} V-cm^2, which corresponds to an electron mean energy of around 7 eV. A reasonable upper limit for the effective E/n is 400 x 10^{-17} V-cm^2 for an extremely fast rising voltage pulse; this condition corresponds to an electron mean energy of about 9 eV.

The dissipation of the input electrical power for

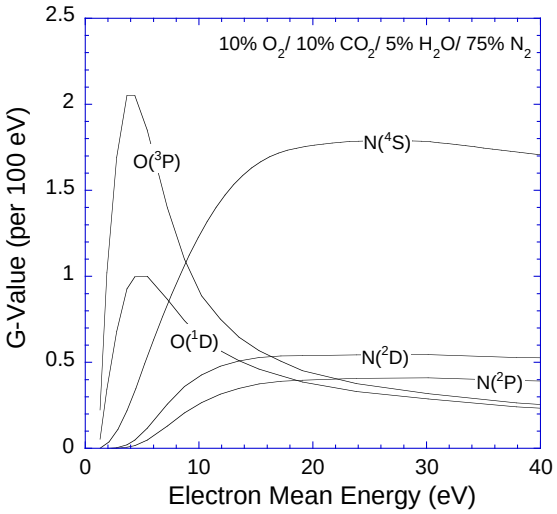

Figure 9. Radical production efficiencies (number of radicals produced per 100 eV of energy input) as functions of the average kinetic energy of the electrons in a plasma for a gas mixture simulating a diesel engine exhaust. The electron mean energy in most electrical discharge reactors is typically between 3 to 6 eV. In this range, a large amount of oxidizing radicals is produced relative to reducing radicals. The ground state oxygen atoms, $O(^3P)$, convert NO to NO_2. The excited oxygen atoms, $O(^1D)$, react with H_2O to produce OH radicals. The OH radicals convert NO and NO_2 to nitrous acid and nitric acid, respectively. High electron mean energies are required to maximize the production of ground state nitrogen atoms, $N(^4S)$, which serve to chemically reduce NO to N_2 and O. The excited nitrogen atoms, $N(^2D)$ and $N(^2P)$, react with the background O_2 to produce NO.

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a gas mixture simulating a diesel engine exhaust is shown in Figure 8. Note that at low electron mean energies (< 5 eV) a large fraction of the input electrical energy is consumed in the vibrational excitation of N_2. Electron mean energies around 5 eV are optimum for the electron-impact dissociation of O_2, which is important for the production of O radicals. To implement the chemical reduction of NO to benign molecules such as N_2 and O_2, the important reducing species is the N atom, which is produced through the electron-impact dissociation of N_2. High electron mean energies are required to efficiently implement the dissociation of N_2. The radical production efficiencies in a plasma for a gas mixture simulating a diesel engine exhaust is shown in Figure 9. The electron mean energy in most electrical discharge reactors is typically between 3 to 6 eV. With an extremely fast rising voltage pulse, a pulsed corona reactor may be able to attain electron mean energies of up to 9 eV. Even for an electron mean energy of 10 eV, a large amount of oxidizing radicals is produced relative to reducing radicals. Electron mean energies much greater than 10 eV are required to maximize the reduction process relative to the oxidation process.

A comparison between modeling and experiment for pulsed corona processing of 100 ppm NO in a simulated diesel engine exhaust is shown in Figure 10. In this case, NO is mostly oxidized to NO_2 and HNO_3.

Our analysis of the plasma chemistry suggests

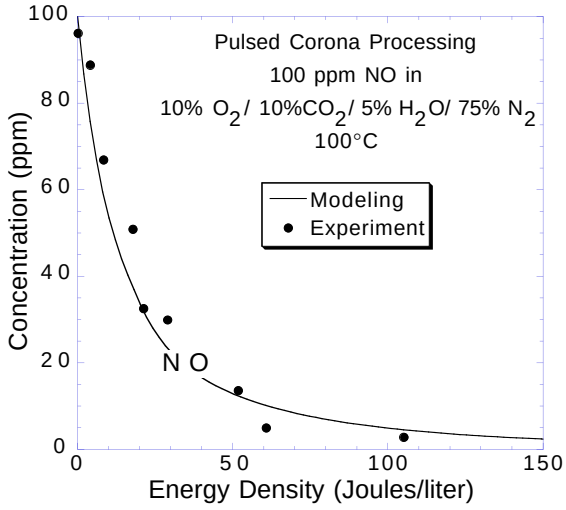

Figure 10. Pulsed corona processing of 100 ppm NO in a gas mixture simulating a diesel engine exhaust at temperature of 100°C and gas flow rate of 65 standard liters per minute. The energy density is in units of Joules per standard liter. The decrease in NO is accomplished mostly via oxidation to NO_2 and HNO_3.

that the best way to optimize NO reduction is to maximize the electron mean energy in the plasma. Our studies have shown that the electron mean energy in discharge reactors is very limited. We therefore considered using an electron beam reactor to achieve the high electron energies in the plasma. In the past, the high capital cost and x-ray hazard associated with conventional electron accelerators have discouraged the use of this technique in small scale applications like engine exhaust treatment. Recently, compact low-energy (≤ 200 keV) electron accelerators have been developed to meet the requirements of industrial applications such as crosslinking of polymer materials and curing of inks, coatings and adhesives. Special materials have also been developed to make the window thin and rugged. Some of these compact electron beam sources are already commercially available. One example [3] of such a source is shown in Figure 11.

Using dilute mixtures of NO in N_2, we have measured the specific energy consumption for electron-impact dissociation of N_2 in an electron beam reactor [4]. We have verified that electron beam processing is much more efficient compared to electrical discharge processing in dissociating N_2. The corresponding specific energy cost for NO reduction by electron beam processing is around 40 eV per NO molecule. With this data we estimated the maximum amount of NO reduction in units familiar to the diesel industry. Figure 12 shows the conversion from "eV per NO molecule" to "grams of NO per brake-horsepower-hour". The maximum amount of NO reduction is directly proportional to the total number of N_2 dissociations that can be achieved in the plasma. As shown in Figure 12, the maximum NO reduction is around 2 grams NO per brake-horsepower-hour at a power consumption ratio of 10%, or 1 gram NO per brake-horsepower-hour at a power consumption ratio of 5%. The amount of NO re-

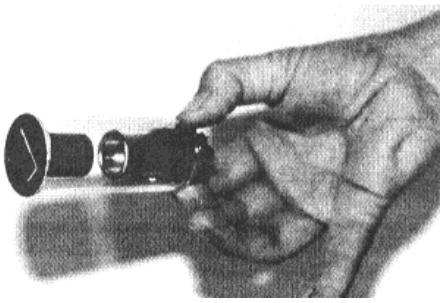

Figure 11. A compact electron beam source developed jointly by Lawrence Livermore National Laboratory and American International Technologies, Inc. The tube is a low cost alternative to large electron beam processing systems and does not require extensive x-ray shielding. The key technical challenge has been the development of a reliable thin membrane window capable of transmitting electron current densities of several milliamperes per square centimeter with 90% efficiency at 50 kilovolts. This device won an R&D 100 award in 1995. See R&D Magazine, September 1995, p. 51 and p. 55.

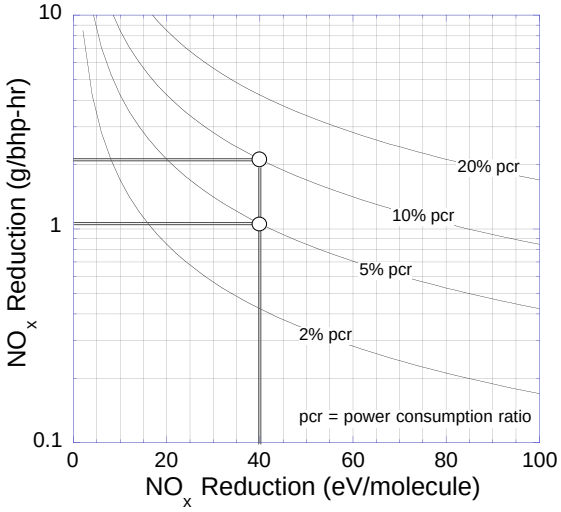

Figure 12. Conversion from "eV per NO molecule" to "grams of NO per brake-horsepower-hour". The value corresponding to 40 eV per molecule is the projected estimate of achievable NO_x reduction using an electron beam reactor in the absence of catalysts or additives. This estimate is based on the total number of N_2 dissociations that can be achieved in the plasma created by electron beam processing.

	Specific Cross Sections ($\times 10^{-17}$ cm^2)			
	N^+	$N(^4S)$	$N(^2D)$	$N(^2P)$
Dissociation Channel				
$N^+ + N(^4S; ^2D)$	6.1	3.05	3.05	
$N(^4S) + N(^4S)$		~0.2		
$N(^4S + N(^2D)$		7.7	10.5	
$N(^4S) + N(^2P)$		7.2		7.2
Total	6.1	18.2	13.6	7.2
Quantum Yield	0.135	0.40	0.30	0.165

Figure 13. Products of the dissociation of N_2 by the impact of 100 eV electrons. From E. C. Zipf, et al., "The Excitation and Collisional Deactivation of Metastable N(^{2}P) Atoms in Auroras", Journal of Geophysical Research, Vol. 85, pp. 687-694 (1980).

duction obtained by electron beam processing represents the maximum NO reduction that can be achieved in any type of atmospheric-pressure non-thermal plasma reactor.

We have encountered complications coming from the electron-impact dissociation of N_2. Dissociative excitation of N_2 contributes a large fraction to the total N_2 dissociation. A significant species produced by dissociative excitation of N_2 is the excited N atom, N(^{2}D). The N(^{2}D) species is a very long lived metastable species. Almost half of the total N radicals produced are in the excited metastable state [5], as shown in Figure 13. The rate constants characterizing the interaction of the metastable species N(^{2}D) with various gases [6] are shown in Figure 14. There are two competing reactions involving the N(^{2}D) metastable species:

NO reduction: $N(^2D) + NO \rightarrow N_2 + O$
NO production: $N(^2D) + O_2 \rightarrow NO + O$

With 1000 ppm NO and 10% O_2, the N(^{2}D) species is ten times more likely to react with O_2 than with NO. This means that N(^{2}D) is consumed in the production of NO rather than in the reduction of NO. Whereas the reaction of ground state N atoms, N(^{4}S), with O_2 can proceed only at very high temperatures, the reaction of excited N atoms, N(^{2}D), with O_2 can proceed even at room temperature. Since almost half of the total N atoms produced in the plasma are in the excited state, the reduction of NO by the ground state N atoms is almost completely counterbalanced by the production of NO by the excited N atoms. What is left in terms of NO

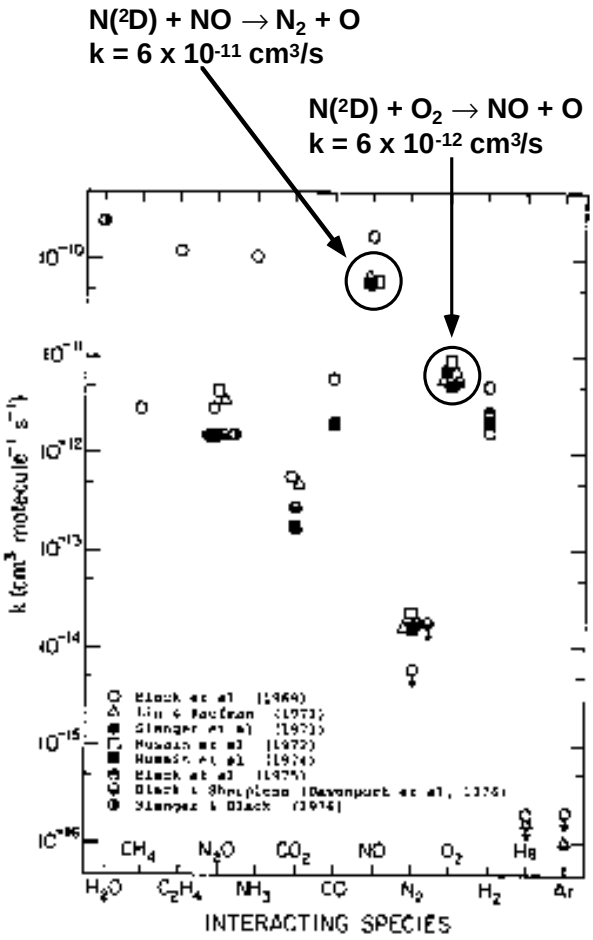

Figure 14. Rate constants characterizing the interaction of the metastable species N(^{2}D) with various gases. From K. Schofield, "Critically Evaluated Rate Constants for Gaseous Reactions of Several Electronically Excited Species", Journal of Physical and Chemical Reference Data, Volume 8, pp. 723-798 (1979). With 1000 ppm NO and 10% O_2, the N(^{2}D) species is ten times more likely to react with O_2 than with NO. NO reduction is thus easily overwhelmed by NO production in the presence of O_2.

reactions is the oxidation reaction

$$O + NO + M \rightarrow NO_2 + M.$$

Calculations showing the effect of the metastable species $N(^2D)$ on electron beam processing of 1000 ppm NO in a 90% N_2/10% O_2 mixture is shown in Figure 15. These calculations have been validated by comparison with experiments performed at the Japan Atomic Energy Research Institute (JAERI) [7]. A comparison of our modeling with JAERI experiments for electron beam processing of 500 ppm NO in 97% N_2/3% O_2 is shown in Figure 16. This comparison suggests that it is indeed the $N(^2D)$ that is responsible for the deleterious effect of O_2 in electron beam processing.

We have suggested electron beam processing as a viable technique for maximizing the production of N atoms which are needed for NO reduction. We have discovered, however, complications in the process due to the abundance of N atoms in the electronically excited state. These metastable excited species react rapidly with O_2 to produce NO, and counteract the NO reduction process achieved by the ground state N radicals. A critical problem now facing us is whether there are ways to inhibit the $N(^2D)$ + O_2 reaction. We next investigated whether the presence of H_2O and CO_2 could quench the metastable N atoms.

The rate constant for interaction of $N(^2D)$ with H_2O is large, as shown in Figure 17. This reaction produces one reducing radical and one oxidizing radical:

$$N(^2D) + H_2O \rightarrow NH + OH.$$

With 10% O_2 and 5% H_2O, $N(^2D)$ is 20 times more likely to react with H_2O than with O_2. The presence of H_2O can thus easily inhibit the reaction $N(^2D) + O_2$. NH is a reducing radical. It can reduce NO via

$$NH + NO \rightarrow N_2 + OH$$

However, in the presence of O_2, NH can react to produce NO via

$$NH + O_2 \rightarrow NO + OH.$$

What is the probability that NH will reduce NO rather than produce NO?

The rate constants [8] for the reactions of NH with NO and O_2 are shown in Figure 18. Note

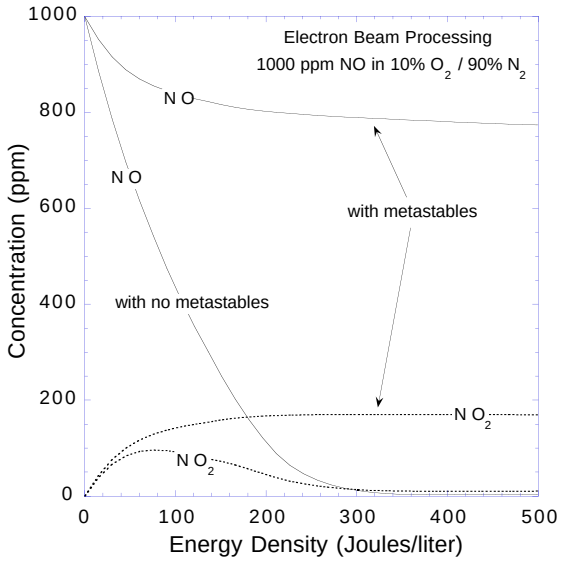

Figure 15. Calculations showing the effect of metastable species $N(^2D)$ on electron beam processing of 1000 ppm NO in a 90% N_2/10% O_2 mixture.

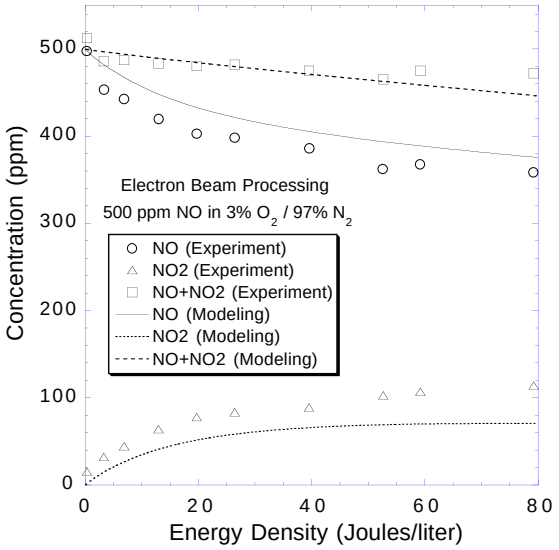

Figure 16. Comparison of LLNL modeling with JAERI experiments for electron beam processing of 500 ppm NO in a 97% N_2/3% O_2 mixture.

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that at 100°C, with 1000 ppm NO and 10% O_2, the NH radical is 30 times more likely to react with NO than with O_2.

The above analysis shows that the production

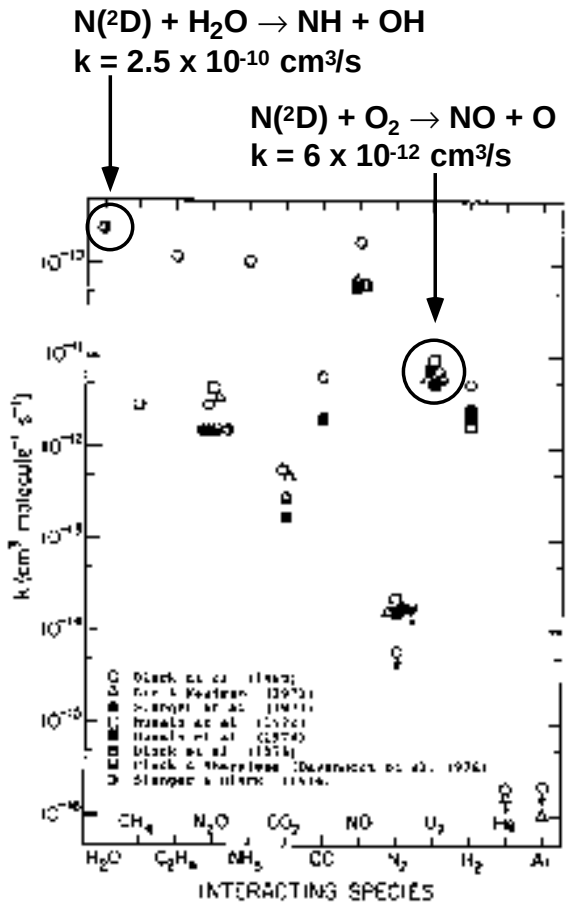

Figure 17. The rate constant for interaction of $N(^2D)$ with H_2O is large. With 10% O_2 and 5% H_2O, $N(^2D)$ is 20 times more likely to react with H_2O than with O_2. The presence of H_2O can thus easily inhibit the reaction $N(^2D) + O_2$.

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of NO from $N(^2D)$ can be prevented by the presence of H_2O. The presence of H_2O however could produce a significant amount of OH radicals in the plasma. The OH radicals could scavenge the NO molecules via

$$NO + OH + M \rightarrow HNO_2 + M.$$

The species HNO_2 is unstable at temperatures above 100°C and decomposes

$$2HNO_2 \rightarrow NO + NO_2 + H_2O$$

Thus although the production of NO from $N(^2D)$ can be prevented by the presence of H_2O, one is still left with a competition between oxidation and reduction. The reduction reaction is the reaction of NO with NH; the oxidation reaction is the reaction of NO with OH. The next question is: what is the probability that NO will react with

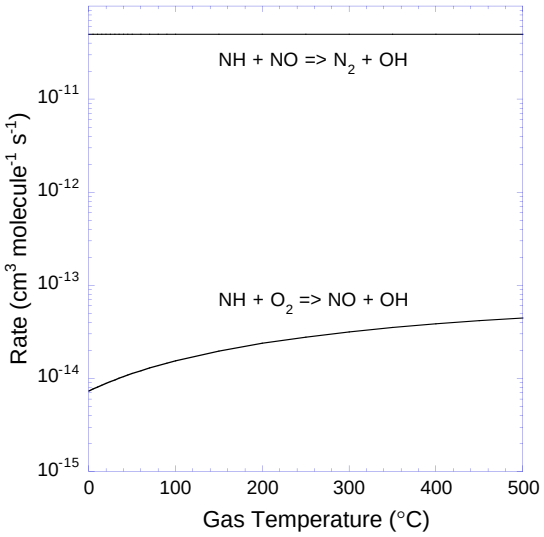

Figure 18. Rate constant for the reactions of NH with NO and O_2. At 100°C, with 1000 ppm NO and 10% O_2, the NH radical is 30 times more likely to react with NO than with O_2. The NH radical therefore will reduce, instead of produce, NO.

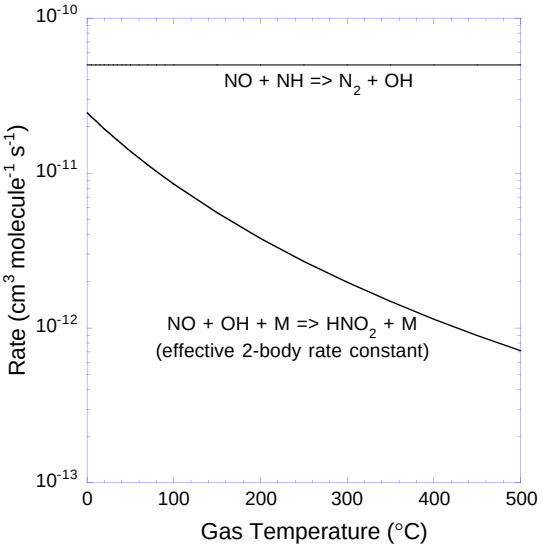

Figure 19. Rate constants for the reactions of NO with NH and OH. At 100°C, NO is 6 times more likely to be reduced by NH rather than oxidized by OH, provided there are about the same number of NH as OH.

NH rather than with OH?

The rate constants [8] for the reactions of NO

with NH and OH are shown in Figure 19. Note that at 100°C, NO is 6 times more likely to react with NH than with OH, provided there are about the same number of NH as OH. Plasma processing in the presence of H_2O will tend to produce a large amount of OH radicals. There are two options to minimize the oxidation of NO by OH radicals: (1) scavenge the OH radicals with a species other than NO, and (2) decrease the production of OH radicals. The first option may be difficult. Based on our understanding of OH production mechanisms [2], we believe the second option may be possible. The contributions of various processes to the production of OH are shown in Figure 20.

In discharge reactors for which the electron mean energy is low, the OH radicals are produced via three types of reactions:

1. Electron attachment:

$$e + H_2O \rightarrow H^- + OH$$

2. Direct dissociation by electron impact:

$$e + H_2O \rightarrow e + H + OH$$

3. Dissociation by $O(^1D)$:

$$O(^1D) + H_2O \rightarrow 2\ OH$$

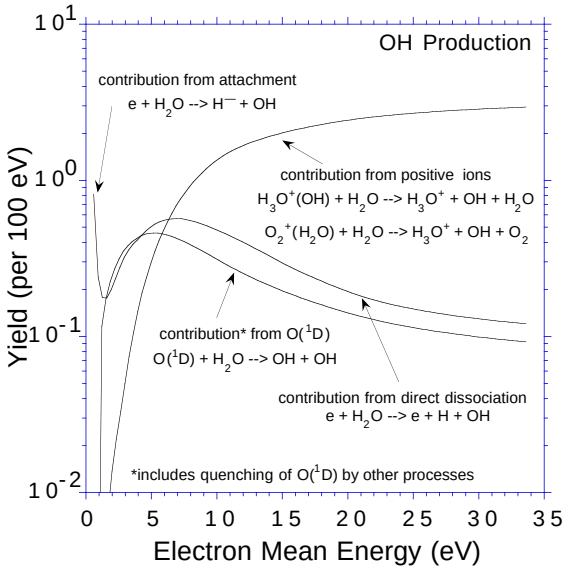

Figure 20. Contributions of various processes to the production of OH as a function of the electron mean energy in the plasma for a gas mixture of 5% O_2, 10% H_2O, 15% CO_2 and 70% N_2. In electron beam processing, the OH radicals come mainly from the positive ions reacting with H_2O.

In electron beam reactors, the OH radicals come mainly from the positive ions reacting with H_2O. The sequence of fast steps are as follows:

Electron-impact ionization:

$$e + O_2 \rightarrow 2e + O_2^+$$

and similar ionization processes to produce molecular ions N_2^+, H_2O^+, CO_2^+

Electron-impact dissociative ionization:

$$e + O_2 \rightarrow 2e + O + O^+$$

and similar dissociative ionization processes to produce N^+, H^+

Charge transfer reactions to form additional O_2^+ ions, such as:

$$N_2^+ + O_2 \rightarrow N_2 + O_2^+$$

Formation of water cluster ions:

$$O_2^+ + H_2O \rightarrow O_2^+(H_2O)$$

Dissociative reactions of water cluster ions to form OH:

$$O_2^+(H_2O) + H_2O \rightarrow H_3O^+ + O_2 + OH$$

$$O_2^+(H_2O) + H_2O \rightarrow H_3O^+(OH) + O_2$$

followed by

$$H_3O^+(OH) + H_2O \rightarrow H_3O^+ + H_2O + OH$$

It may be possible to decrease OH production by neutralizing the positive ions before they have a chance to react with H_2O. We have some ideas of how to implement this. We will try to verify these ideas with experiments.

Summary

We have suggested electron beam processing as a viable technique for maximizing the production of N atoms which are needed for NO reduction. We have encountered, however, complications in the process due to the abundance of N atoms in the electronically excited state. These metastable excited species react rapidly with O_2 to produce NO, and counteract the NO reduction process achieved by the ground state N radicals. The production of NO from $N(^2D)$ can be prevented by the presence

of H_2O. The presence of H_2O however can introduce a large amount of OH radicals that could oxidize NO. We understand how the OH radicals are produced and it may be possible to decrease the OH production in an electron beam reactor. If this succeeds then we may be able to demonstrate chemical reduction of 2 grams NO_x per brake-horsepower-hour at a power consumption ratio of around 10%.

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References

[1] B. Eliasson and U. Kogelschatz, "Electron Impact Dissociation in Oxygen", *J. Phys. B: At. Mol. Phys.* **19**, 1241 (1986).

[2] B. M. Penetrante, "Plasma Chemistry and Power Consumption in Non-Thermal DeNO_x", in *Non-Thermal Plasma Techniques for Pollution Control - Part A: Overview, Fundamentals and Supporting Technologies*, B. M. Penetrante and S. E. Schultheis, Eds. (Springer-Verlag, Berlin Heidelberg, 1993) pp. 65-90.

[3] "Low-Cost Sealed-Tube Electron Beam Gun", in *R&D Magazine*, September 1995, p. 51.

[4] B. M. Penetrante, et. al., "Electron Impact Dissociation of Molecular Nitrogen in Atmospheric Pressure Non-Thermal Plasma Reactors", *Appl. Phys. Lett.* (November 20, 1995).

[5] E. C. Zipf, et al., “The Excitation and Collisional Deactivation of Metastable N(^{2}P) Atoms in Auroras”, *J. Geophys. Res.* **85**, 687 (1980).

[6] K. Schofield, “Critically Evaluated Rate Constants for Gaseous Reactions of Several Electronically Excited Species”, *J. Phys. Chem. Ref. Data* **8**, 723 (1979).

[7] O. Tokunaga et al., "Radiation Treatment of Exhaust Gases - I. Oxidation of NO and Reduction of NO_2", *Int. J. Appl. Radiat. Isotopes* **29**, 81 (1978).

[8] R. Atkinson et al., "Evaluated Kinetic and Photochemical Data for Atmospheric Chemistry: Supplement IV", *J. Phys. Chem. Ref. Data* **21**, 1125 (1992).